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LETTER TO THE EDITOR

***Herschel*/HIFI observations of Mars: First detection of O₂ at submillimetre wavelengths and upper limits on HCl and H₂O₂^{*}**

P. Hartogh¹, C. Jarchow¹, E. Lellouch², M. de Val-Borro¹, M. Rengel¹, R. Moreno², A. S. Medvedev¹, H. Sagawa^{1,3}, B. M. Swinyard⁴, T. Cavalié¹, D. C. Lis⁵, M. I. Błęcka⁶, M. Banaszkiewicz⁶, D. Bockelée-Morvan², J. Crovisier², T. Encrenaz², M. Küppers⁷, L.-M. Lara⁸, S. Szutowicz⁶, B. Vandenbussche⁹, F. Bensch¹⁰, E. A. Bergin¹¹, F. Billebaud¹², N. Biver², G. A. Blake⁵, J. A. D. L. Blommaert⁹, J. Cernicharo¹³, L. Decin^{9,14}, P. Encrenaz¹⁵, H. Feuchtgruber¹⁶, T. Fulton¹⁷, T. de Graauw^{18,19,20}, E. Jehin²¹, M. Kidger²², R. Lorente²², D. A. Naylor²³, G. Portyankina²⁴, M. Sánchez-Portal²², R. Schieder²⁵, S. Sidher⁴, N. Thomas²⁴, E. Verdugo²², C. Waelkens⁹, N. Whyborn²⁰, D. Teyssier²², F. Helmich¹⁸, P. Roelfsema¹⁸, J. Stutzki²⁵, H. G. LeDuc²⁶, and J. A. Stern²⁶

(Affiliations are available on page 5 of the online edition)

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ABSTRACT

We report on an initial analysis of *Herschel*/HIFI observations of hydrogen chloride (HCl), hydrogen peroxide (H₂O₂), and molecular oxygen (O₂) in the Martian atmosphere performed on 13 and 16 April 2010 ($L_s \sim 77^\circ$). We derived a constant volume mixing ratio of 1400 ± 120 ppm for O₂ and determined upper limits of 200 ppt for HCl and 2 ppb for H₂O₂. Radiative transfer model calculations indicate that the vertical profile of O₂ may not be constant. Photochemical models determine the lowest values of H₂O₂ to be around $L_s \sim 75^\circ$ but overestimate the volume mixing ratio compared to our measurements.

Key words. planets and satellites: atmospheres – radiative transfer – submillimeter: general – molecular processes

1. Introduction

Hydrogen chloride (HCl) is a reservoir of chlorine species and plays an important role in the atmospheric chemistry of Venus and Earth. Its detection by ground-based infrared spectroscopy (Iwagami et al. 2008) and space borne UV stellar/solar occultation observations by SPICAV/SOIR on Venus Express (Bertaux et al. 2009) provide mid atmospheric mixing ratios between 0.1 and 1 ppm in the Venusian atmosphere. Submillimetre wave observations of HCl in the Earth atmosphere have long been performed from an airplane (Crewell et al. 1994; Wehr et al. 1995). The derived relative abundances are ~ 2 orders of magnitude smaller than in Venus ($\sim 1\text{--}3$ ppb). In the Martian atmosphere HCl has not been found yet. Its detection would be an indication of present volcanic activity on Mars (Wong et al. 2003; Encrenaz et al. 2004). Krasnopolsky et al. (1997) presented a stringent upper limit of 2 ppb from high-resolution ground-based observations of Mars.

The situation is somewhat different for hydrogen peroxide (H₂O₂). It was detected for the first time in 2003 by Clancy et al. (2004) and Encrenaz et al. (2004) in the Martian atmosphere. The observed abundance varied between 20 and 40 ppb, consistent with photochemical model calculations (e.g.; Krasnopolsky 1993; Atreya & Gu 1994; Nair et al. 1994) for the northern fall season ($L_s = 206^\circ$). H₂O₂ may also be produced by electrostatic discharge reactions during dust storms, in dust devils, or during

normal saltation (Atreya et al. 2006). Near the surface, the concentration could exceed 200 times that produced by photochemistry alone, enough for condensation and precipitation of H₂O₂ to occur. In its solid phase on the surface, it may be responsible for scavenging organic material from Mars and/or present a sink of methane such that a larger source is required to maintain its steady-state abundance (e.g. Mumma et al. 2009).

Oxygen was claimed to be detected for the first time in the Martian atmosphere (in addition to water) by Very (1909). It took almost 60 years until Belton & Hunten (1968) tentatively confirmed the detection of O₂ in the oxygen A band (around 763 nm) with a mixing ratio of 2600 ppm or less. They claimed that the CO/O₂ ratio was two, consistent with the assumption that both gases were produced by the decomposition of CO₂. By performing observations of the same wavelength range, Barker (1972) and Carleton & Traub (1972) found only 1300 ppm of O₂. Since Kaplan et al. (1969) had in the meanwhile reported a reliable measurement of 800 ppm of CO, they concluded that there was an additional source of O₂ namely most likely water. Molecular oxygen is a non-condensable species in the Martian atmosphere. The pressure of the Martian atmosphere oscillates annually by about a third due to the condensation and sublimation of CO₂, i.e. this variation should also appear in the O₂ volume mixing ratio. England & Hrubec (2004) re-analyzed the Viking lander data and found variations from 2500 to 3300 ppm. They point out that the 1300 ppm published by Owen et al. (1977) are not based on Viking measurements, but on the ground-based data cited above and claim that the amount of 3000 ppm is high enough to directly extract oxygen for use as

^{*} *Herschel* is an ESA space observatory with science instruments provided by European-led Principal Investigator consortia and with important participation from NASA.

Table 1. HIFI observations of HCl, H₂O₂ and O₂ in Mars.

OD	Obs. ID	Integration time [s]	UT start date	Molecule	Transition	Sideband	Frequency [GHz]	Beam size ["]
334	1342194690	9289	2010-04-13 06:39:28	O ₂	5,4 → 3,4	LSB	773.840	27.4
				C ¹⁷ O	7 → 6	USB	786.281	27.0
334	1342194689	2297	2010-04-13 05:59:40	O ₂	5,4 → 3,4	USB	773.840	27.4
337	1342194756	2505	2010-04-16 14:53:08	H ₂ O ₂	5 → 4	USB	1847.123	11.5
				CO	16 → 15	LSB	1841.346	11.5
337	1342194755	3746	2010-04-16 13:48:47	HCl	3,4 → 2,4	USB	1876.211	11.3
					3,3 → 2,4		1876.218	11.3
					3,2 → 2,1		1876.223	11.3
					3,3 → 2,2		1876.223	11.3
					3,4 → 2,3		1876.227	11.3
					3,5 → 2,4		1876.227	11.3
					3,3 → 2,3		1876.235	11.3
					3,2 → 2,2		1876.240	11.3
					3,2 → 2,3		1876.252	11.3

a propellant for sample or crew return as well as for the breathing of astronauts (England & Hrubes 2001).

The observations of the HCl, H₂O₂, and O₂ in the Martian atmosphere are part of the *Herschel* key programme “Water and related chemistry in the solar system” (Hartogh et al. 2009). This paper describes the observations and data analysis and provides the volume mixing ratios of the gases and their upper limits.

2. *Herschel*/HIFI observations

The set of HIFI observations was carried out between 11 and 16 April 2010 corresponding to $L_s = 75.8^\circ$ to 78, including spectral line surveys of bands 1a – band 6b (band 5b was not available because of technical problems) and dedicated line observations of carbon monoxide and its isotopes, and water and its isotopes. The telescope was used in a dual-beam switch mode with the source placed alternatively in one of the two beams and cold sky in the other beam, a method that yields very flat baselines (de Graauw et al. 2010; Roelfsema et al. 2010). A summary of the observations is presented in Table 1. We note that Mars was not resolved, since its apparent diameter changed from 8.1 to 8.3'' during the observations. Thus, our observations provide globally averaged quantities. The HCl multiplet at 1876 GHz and the H₂O₂ doublet at 1847 GHz were observed on operational day (OD) 337 with 3746 and 2505 s integration times, respectively, both in the upper sideband (USB) (see Table 1). The O₂ rotational transition at 774 GHz was observed twice on OD 334, once in the upper sideband with 2297 s and once in the lower sideband (LSB) with 9289 s as a side product of a dedicated line observation in the USB. The first set of data was available about a week after the observations and was processed with the standard HIPE v3.0.1 modules (Ott 2010) up to level 2. This data set remained incomplete at the start of our study, for instance the data of the high resolution spectrometer (HRS) was only partly available and pointing products therein had no entries, thus, we analyzed only the wide band spectrometer (WBS) data. This has no impact on the accuracy of the results presented in this paper, although HRS data will be useful for future work including the retrieval of vertical profiles. Since the absolute flux calibration in the data set we obtained from the *Herschel* Science Archive was still in progress, the line-to-continuum ratio was analyzed rather than the absolute brightness temperatures, as is standard for ground-based and other *Herschel* observations (Lellouch et al. 2010; Swinyard et al. 2010).

3. Analysis and discussion

Compared to cometary observations of HIFI (Hartogh et al. 2010b; de Val-Borro et al. 2010), the baseline ripple on the Mars observations is rather large, (as frequently experienced by ground-based telescope observations of planets), because of its strong continuum emission. While in the cometary case the baseline ripple has been removed with a polynomial fit, in the case of Mars we determined the baseline frequencies by a normalized periodogram according to Lomb (1976) and subtracted them from the original spectrum. This was applied separately for horizontal and vertical polarization. After removal of the baseline ripple, both polarizations were averaged. In the case of O₂ observations, we found that the line strengths in both sidebands were the same, and we therefore averaged the spectra obtained in both sidebands.

The observed spectral lines were modeled using a standard radiative transfer code: Mars was assumed to be a perfect sphere surrounded by a set of a hundred concentric atmospheric layers each of 1 km thickness (compare Rengel et al. 2008). Within each layer, the atmospheric temperature, pressure, and volume mixing ratio of carbon monoxide were assumed to be constant. The surface continuum emission was modeled as black-body emission using a temperature distribution falling off towards the edge of the apparent disk according to $T(\alpha) = T_0 \times (1 - 0.2 \times (1 - \cos(\alpha)))$, with α running from 0–90 Å across the apparent disk (see also Cavalié et al. 2008). The disk-averaged emission was obtained by integrating over the apparent disk using sixty four concentric rings distributed unevenly over the disk and the limb region. The variation in the path lengths through the atmosphere were fully taken into account when calculating the radiation transfer of each ring. In our model, the total continuum flux emitted by the surface depends purely on the choice of the temperature T_0 , which defines the temperature scale for the temperature profile to be retrieved. We adjusted T_0 in such a way to match exactly the total flux of about 4230 Jy predicted by the “Mars continuum model” provided by Lellouch & Amri (2008).

The absorption coefficients of the spectral lines were calculated using the JPL spectral line catalog using the terrestrial isotopic ratios. Pressure broadening coefficients for HCl and H₂O₂ were available only for air, while they have been measured in the laboratory in a CO₂ atmosphere for O₂. Most lab measurements display greater pressure broadening in a CO₂ atmosphere. Its impact on the determination of upper limits is small. A 50%

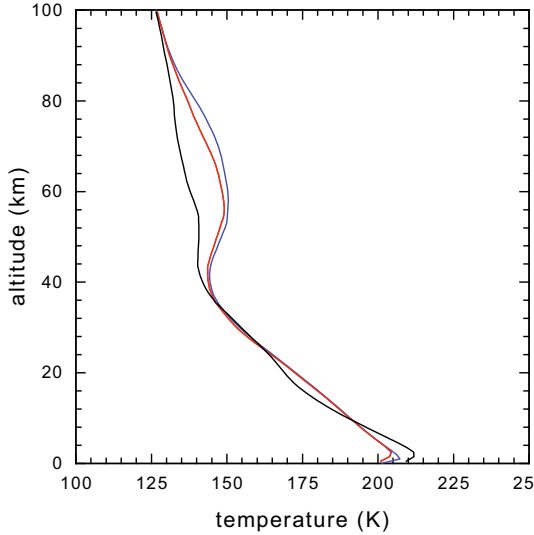


Fig. 1. Temperature profiles predicted by EMCD (blue) (Forget et al. 1999; Lewis et al. 1999), MAOAM (red) (Hartogh et al. 2005; Medvedev & Hartogh 2007), and retrieved vertical profile from simultaneous observations of ¹³CO and C¹⁸O.

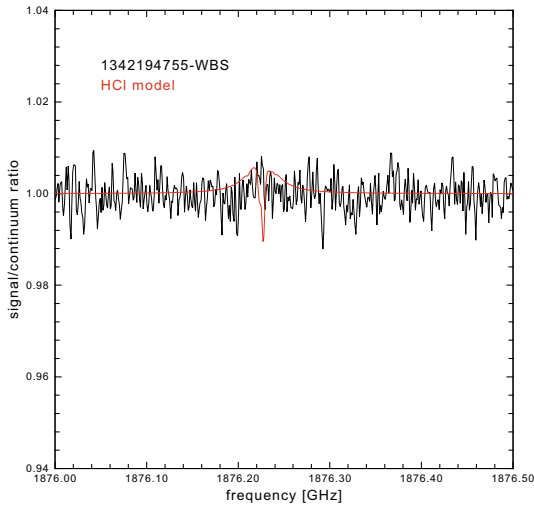


Fig. 2. Observation of HCl centered around 1876 GHz and inserted model calculation (red) for a constant volume mixing ratio of 300 ppt.

increase in the pressure broadening coefficient leads to an increase in the upper limit of 10–20%.

For the retrieval of the mean volume mixing ratio of the three molecules, we applied the temperature profile derived from HIFI observations of ¹³CO and ¹²C¹⁸O during OD 334 (Hartogh et al. 2010a) shown in Fig. 1.

3.1. HCl

Figure 2 shows the result of the 3746 s integration time on the 1876 GHz H³⁵Cl line. We have inserted a modeled spectrum of HCl assuming a constant volume mixing ratio of 300 ppt. HCl was obviously not detected. If we define a line amplitude of 2σ as the upper limit, we derive 200 ppt for HCl. This is one order of magnitude lower than the upper limit derived by (Krasnopolsky et al. 1997) from IR observations. We found no evidence of recent volcanic activity or outgassing from a hot spot on Mars. Nevertheless, the absence of HCl does not preclude extant Martian volcanic activity.

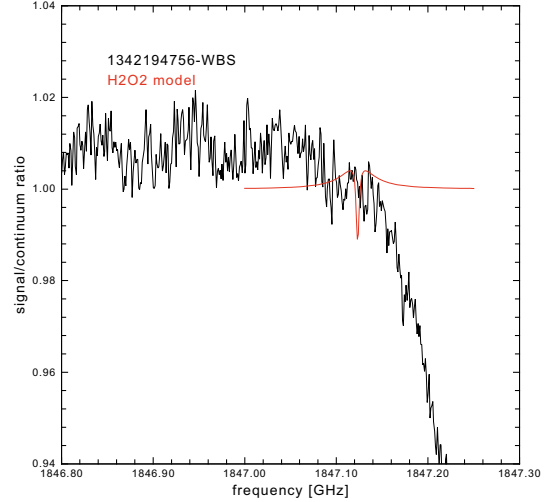


Fig. 3. Observation of H₂O₂ spectrum at 1847 GHz in the upper sideband and inserted model calculation (red) for a constant volume mixing ratio of 3 ppb. The strong absorption feature is CO (16–15) in the lower sideband.

3.2. H₂O₂

Figure 3 shows the result of the H₂O₂ observation on 1847 GHz in the upper sideband. The integration time was 2505 s. The strong absorption feature is the CO (16–15) line. Since the line is in the lower sideband centered around 1841 GHz, it does not absorb any features of the H₂O₂ line. We did not detect any H₂O₂. A modeled H₂O₂ spectrum with a constant volume mixing ratio of 4 ppb has been inserted into the measured spectrum. We deduced a 2-σ upper limit of less than 3 ppb of H₂O₂. At first glance, this value seems far too low taking into account former observations providing 20–40 ppb (see Introduction). On the other hand H₂O₂ is connected to the water cycle and its high variability. Krasnopolsky (2009) compared the annual variability of H₂O₂ based on observations and model calculations averaged over ±35° around the subsolar latitude. Unfortunately, no other observation for $L_s = 78^\circ$ is available. The model calculations provided predictions for this season (Krasnopolsky 2006, 2009; Moudeden & McConnell 2007; Lefèvre et al. 2008), but all overestimated the volume mixing ratio compared to our observation. Lefèvre et al. (2008) found about 10 ppb, Moudeden & McConnell (2007) for $L_s = 90^\circ$ about 15 ppm and even the lowest value of ~5 ppb calculated by Krasnopolsky (2009) is above the upper limit of our observation. Nevertheless, the photochemical models predict lowest H₂O₂ values for the season between $L_s = 70^\circ$ and 80° . Water vapour and its photolysis products are subject to solar cycle variations (Hartogh et al. 2010c). A low Lyman-alpha flux (observations were performed shortly after the solar minimum) may be consistent with less than average production of H₂O₂ in the Martian atmosphere and explain a negative deviation from the model values.

3.3. O₂

The upper panel of Fig. 4 shows the HIFI observation of the 774 GHz O₂ line – the first submm detection of O₂ in Mars – and a model fit of a constant volume mixing ratio. The best fit provides a volume mixing ratio of 1400 ± 120 ppm. This value fits within the error limits to the value of 1300 ppm derived in 1972. We investigated the sensitivity of the pressure broadening coefficient to this value. We initially applied

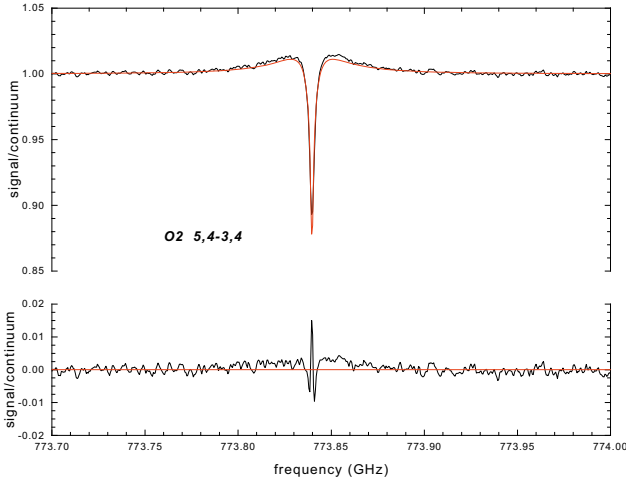


Fig. 4. Observation of O₂ at 774 GHz. The best fit of a constant altitude profile infers a volume mixing ratio of 1400 ± 120 ppm. The lower panel shows the difference between observation and model.

the data from Golubiatnikov & Krupnov (2003) for O₂ in air: $1.62 \text{ MHz hPa}^{-1}$ (half width half maximum, HWHM). Taking into account the higher molecular mass of CO₂ as the main collider compared with air, we multiplied the pressure broadening coefficients in 0.1 hPa steps from 1.1 to 2 and found the best fit of the model to the observation for a factor of 1.2, corresponding to $1.95 \text{ MHz hPa}^{-1}$ (HWHM). We note that the mixing ratio was not found to be very sensitive to these changes, the retrieved value always remaining within the error limits. The pressure broadening factor of 1.2 is smaller than the factor of 1.4 (with CO₂ rather than air being the main collider) for CO that has been found in laboratory measurements (e.g. Dick et al. 2009). The quality of the observation is excellent, the signal-to-noise ratio being higher than 300. Unfortunately, the fit is not optimal. The model underestimates the emission feature and overestimates the depth of the absorption peak. This indicates that the assumption of a constant volume mixing ratio may not be correct. Deviations from the constant profile seem to be positive in the lower and negative in the upper atmosphere. Future work will focus on the vertical profile of O₂.

4. Summary

We have presented initial results for HIFI observations of the Martian atmosphere on HCl, H₂O₂, and O₂. The upper limit of 200 ppt volume mixing ratio determined for HCl is one order of magnitude below the previous value. There is no indication of present volcanic activity. The upper limit to H₂O₂ of 2 ppb is remarkably low compared with former detections. However, this observation is the first one around $L_s = 77^\circ$, a season where photochemical models predict the annual minimum of H₂O₂. Future HIFI observations of H₂O₂ during other solar longitudes will provide additional constraints on photochemical models. The O₂ volume mixing ratio of 1400 ± 120 ppm agrees with former ground-based observations. The assumption of a constant vertical profile does not lead to an optimal fit of the model to the observations. The residuals suggest an oxygen fall off with height. Future work will focus on the retrieval of the vertical O₂ profile.

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- ¹ Max-Planck-Institut für Sonnensystemforschung, 37191 Katlenburg-Lindau, Germany
 - ² LESIA, Observatoire de Paris, 5 place Jules Janssen, 92195 Meudon, France
 - ³ Environmental Sensing & Network Group, NICT, 4-2-1 Nukui-kita, Koganei, Tokyo 184-8795, Japan
 - ⁴ STFC Rutherford Appleton Laboratory, Harwell Innovation Campus, Didcot, OX11 0QX, UK
 - ⁵ California Institute of Technology, Pasadena, CA 91125, USA
 - ⁶ Space Research Centre, Polish Academy of Sciences, Warsaw, Poland
 - ⁷ Rosetta Science Operations Centre, European Space Astronomy Centre, European Space Agency, Spain
 - ⁸ Instituto de Astrofísica de Andalucía (CSIC), Spain
 - ⁹ Instituut voor Sterrenkunde, Katholieke Universiteit Leuven, Belgium
 - ¹⁰ DLR, German Aerospace Centre, Bonn-Oberkassel, Germany
 - ¹¹ Astronomy Department, University of Michigan, USA
 - ¹² Université de Bordeaux, Laboratoire d'Astrophysique de Bordeaux, France
 - ¹³ Laboratory of Molecular Astrophysics, CAB-CSIC, INTA, Spain

- ¹⁴ Sterrenkundig Instituut Anton Pannekoek, University of Amsterdam, Science Park 904, 1098 Amsterdam, The Netherlands
- ¹⁵ LERMA, Observatoire de Paris, France
- ¹⁶ Max-Planck-Institut für extraterrestrische Physik, Giessenbachstraße, 85748 Garching, Germany
- ¹⁷ Bluesky Spectroscopy, Lethbridge, Canada
- ¹⁸ SRON Netherlands Institute for Space Research, Landleven 12, 9747 AD, Groningen, The Netherlands
- ¹⁹ Leiden Observatory, University of Leiden, The Netherlands
- ²⁰ Atacama Large Millimeter/Submillimeter Array, Joint ALMA Office, Santiago, Chile
- ²¹ Institute d'Astrophysique et de Geophysique, Université de Liège, Belgium
- ²² *Herschel* Science Centre, European Space Astronomy Centre, ESA, PO Box 78, 28691 Villanueva de la Cañada, Madrid, Spain
- ²³ Department of Physics and Astronomy, University of Lethbridge, Canada
- ²⁴ Physikalisches Institut, University of Bern, Switzerland
- ²⁵ KOSMA, I. Physik. Institut, Universität zu Köln, Zùlpicher Str. 77, 50937 Köln, Germany
- ²⁶ Jet Propulsion Laboratory, Caltech, Pasadena, CA 91109, USA